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For and on behalf of RWS Group plc

The 8th day of March 2004

**Coextruded, heatsealable and peelable polyester film,  
process for its production and its use**

The invention relates to a coextruded, peelable,  
5 transparent and biaxially oriented polyester film having  
a base layer (B) and at least one top layer (A) applied  
to this base layer (B). The top layer (A) is heatsealable  
and features easy to moderate peelability, in particular  
to CPET trays (CPET = crystalline PET). The heatsealable  
10 and peelable top layer (A) comprises polyester based on  
aromatic and aliphatic acids and aliphatic diols. In  
addition, the top layer (A) comprises a polyester-  
incompatible polymer (anti-PET polymer) in a certain  
concentration. The invention further relates to a process  
15 for producing the film and to its use.

For ready-prepared meals, there are currently double-  
figure growth rates in Europe. The ready-prepared meals  
are transferred to trays after their preparation (cf.  
20 figure 1). A film which is heatsealed to the edge of the  
tray seals the packaging and protects the ready-prepared  
meal from external influences. The ready-prepared meals  
are suitable, for example, for heating in a microwave,  
for heating in a conventional oven or for heating in a  
25 microwave and in a conventional oven. In the latter case,  
the ready-prepared meal and the packaging have to be  
"dual ovenable" (= suitable for microwave and conven-  
tional ovens). As a consequence of the temperatures  
existing in the conventional oven (up to 220 °C),  
30 particularly high demands are made on the packaging  
material (tray and lid film).

Both for the tray and for the lid film, only selected materials can be considered for dual ovenable applications. Typical materials for the trays are in this case CPET, aluminum, cardboard coated with PET or with PET film. In the case of trays made of CPET (cf. figure 1), the thick crystalline CPET layer which is usually pigmented, i.e. filled with particles, provides the stability of the tray, even at the comparatively high temperatures in the conventional oven. Compared to other trays, for example made of amorphous APET, CPET trays have poorer adhesion properties.

In dual ovenable applications, the material used for the lid film is generally PET which is sufficiently dimensionally stable and solid even at 220 °C. Materials such as PP or PE are ruled out from the outset because of their low melting points. The requirements on the lid film are best fulfilled by biaxially oriented polyester film.

When preparing the ready-prepared meal in the oven, the polyester film is removed by hand from the tray shortly before heating or shortly after heating. When this is done, the polyester film must on no account start to tear, start and continue to tear or tear off. The removal of the film from the tray without the film starting or continuing to tear or tearing off is also referred to in the foods industry as peeling. For this application, the polyester film therefore has to be not only heatsealable, but in particular also peelable. For a given material and given overall thickness of the film, the peelability of the film is determined mainly by the properties of the surface layer of the film which is sealed to the tray.

The peelability of films can be determined relatively simply in the laboratory using a tensile strain tester (for example Zwick) (cf. figure 2). For this test, two strips of breadth 15 mm and length approx. 50 mm are first cut out of the polyester film and the tray and sealed together. The sealing layer of the polyester film is formed by the top layer (A) and the sealing layer of the tray by the CPET film. The sealed strips are, as shown in figure 2, clamped into the clips of the tester. The "angle" between the film clamped in the upper clip and the tray strip is 180°. In this test, the clips of the tester are moved apart at a speed of 200 mm/min, and in the most favorable case, the film is fully removed from the tray.

In this test, a distinction is to be drawn between essentially two different mechanisms.

In the first case, the tensile force rises rapidly in the course of the pulling procedure up to a maximum (cf. figure 3a) and then falls directly back to zero. When the maximum force is attained, the film starts to tear, or before delamination, tears off from the tray, resulting in the force falling immediately back to zero. The film is in this case not peelable, since it is destroyed. The behavior of the film can rather be described as a kind of "welding" to the tray. The destruction of the film on removal from the tray is undesired, because this complicates the easy opening of the packaging without tools such as scissors or knives.

In contrast, a peelable film is obtained when the tensile force or the peeling force rises up to a certain value (i.e. up to a certain plateau) and then remains approximately constant over the distance over which the

two strips are sealed together (cf. figure 3b). In this case, the film does not start to tear, but rather can be peeled off as desired from the tray with a low force input.

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The size of the peeling force is determined primarily by the polymers used in the sealing layer (A) (cf. figure 4, polymer 1 and polymer 2). In addition, the size of the peeling force is, dependent in particular on the  
10 heatsealing temperature employed. The peeling force generally rises with the heatsealing temperature. With increasing heatsealing temperature, the risk increases that the sealing layer might lose its peelability. In other words, a film which is peelable when a low  
15 heatsealing temperature is employed loses this property when a sufficiently high heatsealing temperature is employed. This behavior is to be expected in particular in the case of polymers which exhibit the characteristics shown in figure 4 for polymer 1. This behavior which  
20 tends to generally occur but is rather unfavorable for the application has to be taken into account when designing the sealing layer. It has to be possible to heatseal the film in a sufficiently large temperature range without the desired peelability being lost (cf.  
25 polymer 2 in figure 4). In practice, this temperature range is generally from 150 to 220 °C, preferably from 150 to 200 °C and more preferably from 150 to 190 °C.

The heatsealable and peelable layer is applied to the  
30 polyester film in accordance with the prior art, generally by means of offline methods (i.e. in an additional process step following the film production). This method initially produces a "standard polyester film" by a customary process. The polyester film produced  
35 in this way is then coated in a further processing step

in a coating unit offline with a heatsealable and peelable layer. In this process, the heatsealable and peelable polymer is initially dissolved in an organic solvent. The final solution is then applied to the film  
5 by a suitable application process (knife-coater, patterned roller, die). In a downstream drying oven, the solvent is evaporated and the peelable polymer remains on the film as a solid layer.

10 Such an offline application of the sealing layer is comparatively expensive for several reasons. First, the film has to be coated in a separate step in a special apparatus. Second, the evaporated solvent has to be condensed again and recycled, in order thus to minimize  
15 pollution of the environment via the waste air. Third, complicated control is required to ensure that the residual solvent content in the coating is very low.

Moreover, in an economic process, the solvent can never  
20 be completely removed from the coating during the drying, in particular because the drying procedure cannot be of unlimited duration. Traces of the solvent remaining in the coating subsequently migrate via the film disposed on the tray into the foods where they can distort the taste  
25 or even damage the health of the consumer.

Various peelable, heatsealable polyester films which have been produced offline are offered on the market. The polyester films differ in their structure and in the  
30 composition of the top layer (A). Depending on their (peeling) properties, they have different applications. It is customary, for example, to divide the films from the application viewpoint into films having easy peelability (easy peel), having moderate peelability  
35 (medium peel) and having strong robust peelability

(strong peel). The essential quantifiable distinguishing feature between these films is the size of the particular peeling force according to figure 3b. A division is carried out at this point as follows:

5	Easy peelability (easy peel)	Peeling force in the range of from about 1 to 4 N per 15 mm of strip breadth
	Moderate peelability (medium peel)	Peeling force in the range from about 3 to 8 N per 15 mm of strip breadth
	Strong, robust peelability	Peeling force in the range of more than 5 N per 15 mm (strong peel) of strip breadth

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Some sealable PET films are already known.

EP-A-0 035 835 describes a coextruded sealable polyester film to which particles whose average particle size exceeds the layer thickness of the sealing layer are added in the sealing layer to improve the winding and processing performance. The polymer of the sealing film layer is substantially a polyester copolymer which is based on aromatic dicarboxylic acids and also aliphatic diols. The particulate additives form surface elevations which prevent undesired blocking and adhesion of the film to rolls or guides. The selection of particles having a diameter greater than the sealing layer worsens the sealing performance of the film. No information is given in the document on the sealing temperature range of the film. The seal seam strength is measured at 140 °C and is in the range from 63 to 120 N/m (corresponding to from 0.97 to 1.8 N/15 mm of film breadth). There are no indications in the document concerning the peeling

performance of the film with respect to trays made of CPET.

EP-A 0 379 190 describes a coextruded, biaxially oriented  
5 polyester film which comprises a carrier film layer made  
of polyester and at least one sealing film layer made of  
a polyester composition. The sealing film layer may  
comprise aliphatic and aromatic dicarboxylic acids and  
also aliphatic diols. The polymer for the sealing film  
10 layer comprises two different polyesters A and B, of  
which at least one (polyester B) contains aliphatic  
dicarboxylic acids and/or aliphatic diols. The sealing  
energy which is measured between two sealing film layers  
facing each other and joined together (= fin sealing) is  
15 more than  $400 \text{ g}_{\text{force}} \cdot \text{cm}/15 \text{ mm}$  (more than  $4 \text{ N} \cdot \text{cm}/15 \text{ mm}$ ),  
and the sealing film layer may comprise inorganic and/or  
organic fine particles which are insoluble in the  
polyester, in which case the fine particles are present  
in an amount of from 0.1 to 5 % by weight, based on the  
20 total weight of the sealing film layer. In the examples  
of EP-A 0 379 190, organic particles, when they are used  
at all, are used in maximum amounts of 0.3 % by weight.  
Although the film features good peeling properties  
(having plateau character in the peeling diagram [see  
25 above]) with respect to itself (i.e. sealing film layer  
with respect to sealing film layer), there is no  
information about the peeling performance with respect to  
trays made of CPET. In particular, the film of this  
invention is in need of improvement in its producibility  
30 and its processibility (the raw materials tend to  
adhere).

WO A-96/19333 describes a process for producing peelable  
films, in which the heatsealable, peelable layer is  
35 applied inline to the polyester film. In the process,



comparatively small amounts of organic solvents are used. The heatsealable, peelable layer comprises a copolyester for which a) from 40 to 90 mol% of an aromatic dicarboxylic acid, b) from 10 to 60 mol% of an aliphatic dicarboxylic acid, c) from 0.1 to 10 mol% of a dicarboxylic acid containing a free acid group or a salt thereof, d) from 40 to 90 mol% of a glycol containing from 2 to 12 carbon atoms and e) from 10 to 60 mol% of a polyalkyldiol for forming the copolyester were used. The coating is applied to the film from an aqueous dispersion or a solution which contains up to 10 % by weight of organic solvent. The process is restricted with regard to the polymers which can be used and the layer thicknesses which can be achieved for the heatsealable, peelable layer. The maximum achievable layer thickness is specified as 0.5  $\mu\text{m}$ . The maximum seal seam strength is low, and is from 500 to 600 g/25 mm<sup>2</sup>, or [(from 500 to 600)/170] N/15 mm of film breadth.

WO 02/05186 A1 describes a process for producing peelable films, in which the heatsealable, peelable layer is likewise applied inline to the polyester film. In this case, melt-coating is employed, and it is preferably the longitudinally stretched film which is coated with the heatsealable, peelable polymer. The heatsealable, peelable polymer contains polyesters based on aromatic and aliphatic acids, and also based on aliphatic diols. The copolymers disclosed in the examples have glass transition temperatures of below -10 °C; such copolyesters are too soft, which is why they cannot be oriented in customary roll stretching methods (adhesion to the rolls). The thickness of the heatsealable, peelable layer is less than 8  $\mu\text{m}$ . In WO 02/05186 A1, the melt-coating known per se is delimited from the extrusion coating known per se technically and by the viscosity of

the melt. A disadvantage of the process is that only comparatively fluid polymers (max. 50 Pa\*sec) having a low molecular weight can be used. This results in disadvantageous peeling properties of the film. Moreover, the coating rate in this process is limited, which makes the production process uneconomic. With regard to quality, faults are observed in the optical properties of the film which are visible, for example, as coating streaks. In this process, it is also difficult to obtain a uniform thickness of the sealing layer over the web breadth of the film, which in turn leads to nonuniform peeling characteristics.

It is an object of the present invention to provide a coextruded, heatsealable and peelable, biaxially oriented polyester film which features outstanding peeling properties with respect to trays made of CPET. It should no longer have the disadvantages of the prior art films and should in particular have the following features:

- Easy to moderate peelability (easy peel to strong peel) with respect to CPET. The peeling force should be in the range from 1.5 to 8 N per 15 mm, preferably in the range from 2.0 to 7.5 N per 15 mm and more preferably in the range from 2.5 to 7.0 N per 15 mm, of film strip breadth.
- No organic solvent residues are present in the heatsealable and peelable layer.
- The heatsealable and peelable layer, with respect to the CPET tray, has a minimum sealing temperature of 165 °C, preferably 160 °C, in particular 155 °C, and a maximum sealing temperature of generally 220 °C, preferably 200 °C and more preferably 190 °C.
- It is produced employing processes in which no organic solvents are used from the outset.

- The film can be prepared economically. This also means, for example, that stretching processes which are customary in the industry can be used to produce the film. In addition, it should be possible to produce the film at machine speeds of up to 500 m/min which are customary today.
- Good adhesion (greater than 2 N/15 mm of film breadth) between the individual layers of the film is ensured for their practical employment.
- The optical properties of the film are good. This means, for example, low opacity (less than 20 %) and high gloss (> 70 for the sealable side and > 100 for the side opposite the sealable side; each at 20 ° angle of incidence) of the film.
- In the course of the production of the film, it is guaranteed that the regrind can be fed back to the extrusion in an amount of up to 60 % by weight, without significantly adversely affecting the physical (the tensile strain at break of the film in both directions should not decrease by more than 10 %), but in particular the optical, properties of the film.

In addition, care should be taken that the film can be processed on high-speed machines. On the other hand, the known properties which distinguish polyester films should at the same time not deteriorate. These include, for example, the mechanical (the modulus of elasticity of the biaxially stretched films in both orientation directions should be greater than 3000 N/mm<sup>2</sup>, preferably greater than 3500 N/mm<sup>2</sup> and more preferably greater than 4000 N/mm<sup>2</sup>) and the thermal properties (the shrinking of the biaxially stretched films in both orientation directions should not be greater than 3 %, preferably not greater than 2.8 % and more preferably not greater than 2.5 %),

the winding performance and the processibility of the film, in particular in the printing, laminating or in the coating of the film with metallic or ceramic materials.

5 In this context, heatsealable refers to the property of a coextruded, multilayer polyester film which comprises at least one base layer (B) and also comprises at least one top layer (= heatsealable top layer) which can be bonded by means of sealing jaws by applying heat (140 to  
10 220 °C) and pressure (2 to 5 bar) within a certain time (0.2 to 2 sec) to itself (fin sealing), or to a substrate made of a thermoplastic (= lab sealing, in this case in particular with respect to CPET trays), without the carrier layer (= base layer) itself becoming plastic. In  
15 order to achieve this, the polymer of the sealing layer generally has a distinctly lower melting point than the polymer of the base layer. When the polymer used for the base layer is, for example, polyethylene terephthalate having a melting point of 254 °C, the melting point of  
20 the heatsealable layer is generally less than 230°, in the present case preferably less than 210° and more preferably less than 190 °C.

In this context, peelable refers to the property of a  
25 coextruded polyester film which comprises at least one layer (= heatsealable and peelable top layer) which, after heatsealing to a substrate (in this case substantially the CPET tray), can be pulled from the substrate in such a way that the film neither starts to  
30 tear nor tears off. The bond of heatsealable film and substrate breaks in the seam between the heatsealed layer and the substrate surface when the film is removed from the substrate (cf. also Ahlhaus, O.E.: Verpackung mit Kunststoffen [Packing with plastics], Carl Hanser Verlag,  
35 p. 271, 1997, ISBN 3-446-17711-6). When removing the film

heatsealed to a test strip of the substrate in a tensile strain testing instrument at a peeling angle of 180 ° in accordance with figure 2, the tensile strain behavior of the film according to figure 3b is then obtained. When  
 5 peeling off the film from the substrate commences, the force required for this purpose rises, according to figure 3b, up to a certain value (e.g. 4 N/15 mm) and then remains approximately constant over the entire peeling process, but is subject to larger or smaller  
 10 variations (approx. +/-20 %).

This object is achieved by providing a coextruded, transparent, biaxially oriented polyester film comprising a base layer (B) and a heatsealable top layer (A) which  
 15 is peelable with respect to CPET, the heatsealable and peelable top layer (A) consisting of

- a) 70-97 % by weight of polyester and
- b) 3-20 % by weight of a polyester-incompatible polymer (= anti-PET polymer)
- 20 based on the mass of the top layer (A), and
- c) the polyester being composed of 20-95 mol% of units which derive from at least one aromatic dicarboxylic acid and 5-80 mol% of units which derive from at least one aliphatic dicarboxylic acid, the sum of  
 25 the dicarboxylic acid-derived molar percentages being 100, and
- d) the layer thickness of the top layer (A)  $d_A$  being from 1.7 to 6.0  $\mu\text{m}$ .

30

The material of the top layer (A) thus consists predominantly of a polyester and a polyester-incompatible polymer (anti-PET polymer). The polyester is composed of units which are derived from aromatic and aliphatic  
 35 dicarboxylic acids. The units which derive from the

- aromatic dicarboxylic acids are present in the polyester in an amount of 20-95 mol%, preferably 30-90 mol%, more preferably 50-88 mol%. The units which derive from the aliphatic dicarboxylic acids are present in the polyester
- 5 in an amount of 5-80 mol%, preferably 10-70 mol%, more preferably 12-50 mol%, and the molar percentages always add up to 100 %. The diol units corresponding thereto likewise always make up 100 mol%.
- 10 Preferred aliphatic dicarboxylic acids are succinic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, glutaric acid and adipic acid. Particular preference is given to azelaic acid, sebacic acid and adipic acid.
- 15 Preferred aromatic dicarboxylic acids are terephthalic acid, isophthalic acid and 2,6-naphthalenedicarboxylic acid, in particular terephthalic acid and isophthalic acid.
- 20 Preferred diols are ethylene glycol, butylene glycol and neopentyl glycol.

In general, the polyester comprises the following dicarboxylates and alkylenes, based in each case on the

25 total amount of dicarboxylate or total amount of alkylene:

- from 20 to 95 mol%, preferably from 25 to 85 mol% and more preferably from 40 to 78 mol%, of terephthalate,
- 30 • from 0 to 25 mol%, preferably from 5 to 20 mol% and more preferably from 10 to 20 mol%, of isophthalate,
- from 5 to 80 mol%, preferably from 8 to 70 mol% and more preferably from 11 to 47 mol%, of azelate,
- from 0 to 50 mol%, preferably from 0 to 40 mol% and
- 35 more preferably from 0.2 to 30 mol%, of sebacate,

- from 0 to 50 mol%, preferably from 0 to 40 mol% and more preferably from 0 to 30 mol%, of adipate.
- More than 30 mol%, preferably more than 40 mol% and more preferably more than 50 mol%, of ethylene or butylene.

3-20 % by weight, preferably 5-18 % by weight and more preferably 7-15 % by weight, of the top layer material consists of a polymer which is incompatible with polyester (anti-PET polymer).

From 0 to 10 % by weight of the material of the top layer (A) consists of particles, additives, auxiliaries and/or other additives which are customarily used in polyester film technology.

It has been found to be appropriate to produce the main polyester of the top layer (A) from two separate polyesters I and II which are fed to the extruder for this layer as a mixture.

The heatsealable and peelable top layer (A) is distinguished by characteristic features. It has a sealing commencement temperature (= minimum sealing temperature) with respect to CPET trays of not more than 165 °C, preferably not more than 160 °C and more preferably not more than 155 °C, and a seal seam strength with respect to CPET trays of at least 1.5 N, preferably at least 2.0 N, more preferably at least 2.5 N (always based on 15 mm film breadth). The heatsealable and peelable top layer (A), with respect to CPET trays, has a max. sealing temperature of generally 220 °C, preferably 200 °C and more preferably 190 °C, and a film which is peelable with respect to CPET trays is obtained within the entire sealing range. In other words, this film in the 180 °

tensile experiment according to figure 2 provides a curve according to figure 3b.

For the preferred, abovementioned ranges, the peeling results can also be described numerically. According to the present experimental investigations, the peeling results can be correlated together simply by the following relationship between the sealing temperature (in °C) and the peeling force (in N/15 mm)

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$$0.0035 \cdot \delta / ^\circ\text{C} - 3.6 \leq \text{peeling force } F/N \text{ per } 15 \text{ mm} \leq 0.05 \cdot \delta / ^\circ\text{C} - 2.5$$

This relationship is depicted graphically in figure 5 for illustration.

15

The film of the present invention comprises a base layer (B) and at least one top layer (A) according to the invention. In this case, the film has a two-layer structure. In a preferred embodiment, the film has a three- or more than three-layer structure. In the case of the particularly preferred three-layer embodiment, it consists of the base layer (B), the inventive top layer (A) and a top layer (C) on the opposite side to the top layer (A). In a four-layer embodiment, the film comprises an intermediate layer (D) between the base layer (B) and the top layer (A) or (C).

The base layer of the film consists of at least 80 % by weight of thermoplastic polyester. Suitable for this purpose are polyesters of ethylene glycol and terephthalic acid (= polyethylene terephthalate, PET), of ethylene glycol and naphthalene-2,6-dicarboxylic acid (= polyethylene 2,6-naphthalate, PEN), of 1,4-bishydroxymethylcyclohexane and terephthalic acid (= poly-1,4-cyclohexanedimethylene terephthalate, PCDT) and also



of ethylene glycol, naphthalene-2,6-dicarboxylic acid and biphenyl-4,4'-dicarboxylic acid (= polyethylene 2,6-naphthalate bibenzoate, PENBB). Preference is given to polyesters which contain ethylene units and consist, based on the dicarboxylate units, of at least 90 mol%, more preferably at least 95 mol%, of terephthalate or 2,6-naphthalate units. The remaining monomer units stem from other dicarboxylic acids or diols. Advantageously, copolymers or mixtures or blends of the homo- and/or copolymers mentioned can also be used for the base layer (B). (In the specification of the amounts of the dicarboxylic acids, the total amount of all dicarboxylic acids is 100 mol%. Similarly, the total amount of all diols also adds up to 100 mol%.)

Suitable other aromatic dicarboxylic acids are preferably benzenedicarboxylic acids, naphthalenedicarboxylic acids (for example naphthalene-1,4- or 1,6-dicarboxylic acid), biphenyl-x,x'-dicarboxylic acids (in particular biphenyl-4,4'-dicarboxylic acid), diphenylacetylene-x,x'-dicarboxylic acids (in particular diphenylacetylene-4,4'-dicarboxylic acid) or stilbene-x,x'-dicarboxylic acids. Of the cycloaliphatic dicarboxylic acids, mention should be made of cyclohexanedicarboxylic acids (in particular cyclohexane-1,4-dicarboxylic acid). Of the aliphatic dicarboxylic acids, the (C<sub>3</sub>-C<sub>19</sub>)alkanedioic acids are particularly suitable, and the alkane moiety may be straight-chain or branched.

Suitable other aliphatic diols are, for example, diethylene glycol, triethylene glycol, aliphatic glycols of the general formula HO-(CH<sub>2</sub>)<sub>n</sub>-OH where n is an integer from 3 to 6 (in particular propane-1,3-diol, butane-1,4-diol, pentane-1,5-diol and hexane-1,6-diol) or branched aliphatic glycols having up to 6 carbon atoms,

cycloaliphatic, optionally heteroatom-containing diols having one or more rings. Of the cycloaliphatic diols, mention should be made of cyclohexanediols (in particular cyclohexane-1,4-diol). Suitable other aromatic diols correspond, for example, to the formula  $\text{HO-C}_6\text{H}_4\text{-X-C}_6\text{H}_4\text{-OH}$  where X is  $-\text{CH}_2-$ ,  $-\text{C}(\text{CH}_3)_2-$ ,  $-\text{C}(\text{CF}_3)_2-$ ,  $-\text{O}-$ ,  $-\text{S}-$  or  $-\text{SO}_2-$ . In addition, bisphenols of the formula  $\text{HO-C}_6\text{H}_4\text{-C}_6\text{H}_4\text{-OH}$  are also very suitable.

It is particularly advantageous when a polyester copolymer based on terephthalate and small amounts ( $< 5 \text{ mol}\%$ ) of isophthalic acid or based on terephthalate and small amounts ( $< 5 \text{ mol}\%$ ) of naphthalene-2,6-dicarboxylic acid is used in the base layer (B). In this case, the producibility of the film and the optical properties of the film are particularly good. The base layer (B) then comprises substantially a polyester copolymer which is composed predominantly of terephthalic acid and isophthalic acid units and/or terephthalic acid and naphthalene-2,6-dicarboxylic acid units and of ethylene glycol units. The particularly preferred copolyesters which provide the desired properties of the film are those which are composed of terephthalate and isophthalate units and of ethylene glycol units.

The polyesters can be prepared by the transesterification process. In this process, the starting materials are dicarboxylic esters and diols which are reacted with the customary transesterification catalysts such as zinc, calcium, lithium and manganese salts. The intermediates are then polycondensed in the presence of generally customary polycondensation catalysts such as antimony trioxide, titanium oxides or esters, or else germanium compounds. The preparation may equally well be by the direct esterification process in the presence of

polycondensation catalysts. This process starts directly from the dicarboxylic acids and the diols.

5 The film of the present invention has an at least two-layer structure. It then consists of the base layer (B) and the inventive sealable and peelable top layer (A) applied to it by coextrusion.

10 The sealable and peelable top layer (A) applied to the base layer (B) by coextrusion is composed predominantly, i.e. of at least approx. 70 % by weight, of polyesters.

15 According to the invention, the heatsealable and peelable top layer (A) comprises polyesters based on aromatic and aliphatic acids and preferably aliphatic diols. In addition, the top layer (A) comprises a polymer which is incompatible with polyester (anti-PET polymer) in a concentration of 3-20 % by weight.

20 In the preferred embodiment, polyesters are copolyesters or blends of homo- and copolyesters or blends of different copolyesters whose composition is based on aromatic and aliphatic dicarboxylic acids and aliphatic diols.

25 Examples of the aromatic dicarboxylic acids which can be used in accordance with the invention are terephthalic acid, isophthalic acid, phthalic acid and 2,6 naphthalenedicarboxylic acid.

30 Examples of the aliphatic dicarboxylic acids which can be used in accordance with the invention are succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid and sebacic acid.

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Examples of the aliphatic diols which can be used in accordance with the invention are ethylene glycol, 1,3-propanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentenediol, 2,2-dimethyl-1,3-propanediol, diethylene glycol, triethylene glycol and 1,4-cyclohexanedimethanol.

The polyester for the top layer (A) is preferably prepared from two polyesters I and II.

10 The proportion of the polyester I which consists of one or more aromatic dicarboxylates and one or more aliphatic alkylenes in the top layer (A) is from 10 to 60 % by weight. In the preferred embodiment, the proportion of the polyester I is from 15 to 55 % by weight, and in the  
15 particularly preferred embodiment, it is from 20 to 50 % by weight.

In general, the polyester I of the inventive top layer (A) is based on the following dicarboxylates and  
20 alkylenes, based in each case on the total amount of dicarboxylate or total amount of alkylene:

- from 70 to 100 mol%, preferably from 72 to 95 mol% and more preferably from 74 to 93 mol%, of terephthalate,
- 25 • from 0 to 30 mol%, preferably from 5 to 28 mol% and more preferably from 7 to 26 mol%, of isophthalate,
- more than 50 mol%, preferably more than 65 mol% and more preferably more than 80 mol%, of ethylene units.

30 Any remaining fractions present stem from other aromatic dicarboxylic acids and other aliphatic diols, as have already been listed above as main and secondary carboxylic acids for the base layer (B).

Very particular preference is given to those copolyesters in which the proportion of terephthalate units is from 74 to 88 mol%, the corresponding proportion of isophthalate units is from 12 to 26 mol% (the dicarboxylate fractions adding up to 100 mol%) and the proportion of ethylene units is 100 mol%. In other words, they are polyethylene terephthalate/isophthalate.

In a further preferred embodiment, the polyester I consists of a mixture which comprises a copolyester composed of terephthalate, isophthalate and of ethylene units, and an aromatic polyester homopolymer, e.g. a polybutylene terephthalate.

It has been found that in the case that the proportion of polyester I in the top layer (A) is less than 10 % by weight, the producibility of the film by coextrusion technology is made distinctly more difficult, or is no longer guaranteed. The tendency of the film to adhere to certain machine parts, in particular to running metallic rolls in the longitudinal stretching and after the transverse stretching, is particularly high in this case. In contrast, when the proportion of polyester I in the top layer (A) is on the other hand more than 60 % by weight, the peeling performance of the film is strongly impaired. The sealing performance of the film changes in this case from peelable to weldable.

According to the present invention, the proportion of polyester II in the top layer (A) is from 20 to 80 % by weight. In the preferred embodiment, the proportion of polyester II is from 25 to 80 % by weight and in the particularly preferred embodiment, it is from 30 to 80 % by weight.

The polyester II preferably consists of a copolymer of aliphatic and aromatic acid components, in which the aliphatic acid components are from 20 to 90 mol%, preferably from 30 to 70 mol% and more preferably from 35 to 60 mol%, based on the total acid amount of the polyester II. The remaining dicarboxylate content up to 100 mol% stems from aromatic acids, preferably of terephthalic acid and/or of isophthalic acid, and also, among the glycols, from aliphatic or cycloaliphatic or aromatic diols, as have already been described in detail above with regard to the base layer.

In general, the polyester II of the inventive top layer (A) is based at least on the following dicarboxylates and alkylenes, based in each case on the total amount of dicarboxylate or the total amount of alkylene:

- from 20 to 65 mol%, preferably from 30 to 70 mol% and more preferably from 35 to 60 mol%, of azelate,
- from 0 to 50 mol%, preferably from 0 to 45 mol% and more preferably from 0 to 40 mol%, of sebacate,
- from 0 to 50 mol%, preferably from 0 to 45 mol% and more preferably from 0 to 40 mol%, of adipate,
- from 10 to 80 mol%, preferably from 20 to 70 mol% and more preferably from 30 to 60 mol%, of terephthalate,
- from 0 to 30 mol%, preferably from 3 to 25 mol% and more preferably from 5 to 20 mol%, of isophthalate,
- more than 30 mol%, preferably more than 40 mol% and more preferably more than 50 mol%, of ethylene or butylene.

Any remaining fractions present stem from other aromatic dicarboxylic acids and other aliphatic diols, as have already been listed above as main and secondary carboxylic acids for the base layer (B), or else from

hydroxycarboxylic acids such as hydroxybenzoic acid or the like.

5 The presence of at least 10 mol% of aromatic dicarboxylic acid ensures that the polymer II can be processed without adhesion, for example in the coextruder or in the longitudinal stretching.

10 When the proportion of polyester II in the top layer (A) is less than 20 % by weight, the peeling performance of the film is strongly impaired. In this case, the sealing performance of the film changes from peelable to weldable. In contrast, when the proportion of polyester II in the top layer (A) is on the other hand more than  
15 80 % by weight, the producibility of the film by coextrusion technology is made more difficult, or is no longer guaranteed. The tendency of the film to adhere to certain machine parts, in particular to running metallic rolls in the longitudinal stretching and after the  
20 transverse stretching, is particularly high in this case.

The top layer (A) preferably comprises a mixture of the polyesters I and II. Compared to the use of only one polyester with comparable components and comparable  
25 proportions of the components, a mixture has the following advantages:

- The mixture of the two polyesters I and II, from the aspect of the particular glass transition temperatures ( $T_g$ ), is easier to process (to extrude). As  
30 investigations have shown, the mixture of a polymer having a high  $T_g$  (polyester I) and a polymer having a low  $T_g$  (polyester II) has a lesser tendency to adhere in the intake of the coextruder than a single polymer having a correspondingly mixed  $T_g$ .

- The polymer production is simpler, because the number of metering stations available for the starting materials generally cannot be unlimited.
- Moreover, from a practical aspect, the desired peeling properties can be attained more individually with the mixture than when a single polyester is used.
- The addition of particles (see below) is also simpler in the case of polyester I than in the case of polyester II.

Appropriately, the glass transition temperature of polyester I is more than 50 °C. Preference is given to the glass transition temperature of polyester I being more than 55 °C and more preferably more than 60 °C. When the glass transition temperature of polyester I is less than 50 °C, the film cannot be produced in a reliable process. The tendency of the top layer (A) to adhere, for example to rolls, is so high that frequent film breaks, in particular in the longitudinal stretching, have to be expected. When this happens, the film can wind around the rolls in the longitudinal stretching, which can lead to considerable damage to the machine. In the extrusion, such a polyester adheres readily to the metallic walls and thus leads to blockages.

Appropriately, the glass transition temperature of polyester II is less than 20 °C. The glass transition temperature is preferably less than 15 °C and more preferably less than 10 °C. When the glass transition temperature of polyester II is greater than 20 °C, the film has an increased tendency to start to tear or tear off when pulled from the tray, which is undesired.



According to the invention, the heatsealable and peelable top layer (A) comprises a polymer which is incompatible with polyester (anti-PET polymer) in a certain concentration. According to the present invention, the proportion of the polyester-incompatible polymer (anti-PET polymer) is from 3 to 20 % by weight, based on the mass of the top layer (A). In a preferred embodiment, the proportion of the polymer is from 5 to 18 % by weight and in the particularly preferred embodiment, it is from 7 to 15 % by weight, likewise based on the mass of the top layer (A).

Examples of suitable incompatible polymers (anti-PET polymer) are polymers based on ethylene (e.g. LLDPE, HDPE), propylene (PP), cycloolefins (CO), amides (PA) or styrene (PS). In a preferred embodiment, the polyester-incompatible polymer (anti-PET polymer) used is a copolymer. Examples thereof are copolymers based on ethylene ( $C_2/C_3$ ,  $C_2/C_3/C_4$  copolymers), propylene ( $C_2/C_3$ ,  $C_2/C_3/C_4$  copolymers), butylene ( $C_2/C_3$ ,  $C_2/C_3/C_4$  copolymers) or based on cycloolefins (norbornene/ethylene, tetracyclododecene/ethylene copolymers). In one of the particularly preferred embodiments, the polyester-incompatible polymer (anti-PET polymer) is a cycloolefin copolymer (COC). Such cycloolefin copolymers are described generally, for example, in EP-A 1 068 949 or in JP 05-009319, which are incorporated herein by reference.

Among the cycloolefin copolymers, preference is given in particular to those which comprise polymerized units of polycyclic olefins having a norbornene basic structure, more preferably norbornene or tetracyclododecene. Particular preference is given to cycloolefin copolymers (COC) which contain polymerized units of acyclic olefins, in particular ethylene. Very particular preference is

given to norbornene/ethylene and tetracyclo-  
dodecene/ethylene copolymers which contain from 5 to 80  
% by weight of ethylene units, preferably from 10 to 60  
% by weight of ethylene units (based on the mass of the  
5 copolymer).

The cycloolefin polymers generally have glass transition  
temperatures between -20 and 400 °C. Suitable for the  
invention are those cycloolefin copolymers (COC) which  
10 have a glass transition temperature of less than 160 °C,  
preferably less than 120 °C and more preferably less than  
80 °C. The glass transition temperature should preferably  
be above 50 °C, with preference above 55 °C, in  
particular above 60 °C. The viscosity number (decalin,  
15 135 °C, DIN 53 728) is appropriately between 0.1 and  
200 ml/g, preferably between 50 and 150 ml/g. Films which  
comprise a COC having a glass transition temperature of  
less than 80 °C compared to those comprising a COC having  
a glass transition temperature of greater than 80 °C  
20 feature improved optical properties, in particular a  
lower opacity.

The cycloolefin copolymers (COC) are prepared by hetero-  
geneous or homogeneous catalysis with organometallic  
25 compounds and is described in a multitude of documents.  
Suitable catalyst systems based on mixed catalysts of  
titanium or vanadium compounds in combination with  
aluminum organyls are described in DD 109 224, DD 237 070  
and EP-A-0 156 464.

30  
EP-A-0 283 164, EP-A-0 407 870, EP-A-0 485 893 and  
EP-A-0 503 422 describe the preparation of cycloolefin  
copolymers (COC) with catalysts based on soluble  
metallocene complexes. Particular preference is given to  
35 using cycloolefin copolymers prepared with catalysts

which are based on soluble metallocene complexes. Such COCs are commercially obtainable; for example Topas® (Ticona, Frankfurt).

5 When the proportion of the polyester-incompatible polymer (anti-PET polymer) is less than 3 % by weight, based on the weight of the top layer (A), there is no positive influence of the polymer on the removal performance of the film from the tray. When the film is removed from the  
10 tray, it still tends to start to tear or to tear off. Especially at relatively high sealing temperatures (> 160 °C), this effect as a result of the addition of polyester-incompatible polymer (anti-PET polymer) becomes particularly too apparent. Films produced in accordance  
15 with the invention then do not start to tear or tear off on removal from the tray. On the other hand, the proportion of polyester-incompatible polymer (anti-PET polymer) should not exceed 20 % by weight, since the opacity of the film otherwise becomes too high.

20 To improve the handling of the film, the processibility of the film, but in particular also to improve the removal performance of the film from the tray, it is advantageous to further modify the heatsealable and  
25 peelable top layer (A).

This is at best done with the aid of suitable antiblocking agents (particles) which are optionally added to the sealing layer and in such amounts that the  
30 removal performance of the film from the tray is further improved, blocking of the film is prevented and the processing performance of the film is optimized.

It has been found to be advantageous for at least the top  
35 layer (A) to comprise particles in a certain size, in a

certain concentration and in a certain distribution. In addition, mixtures of two and more different particle systems or mixtures of particle systems in the same chemical composition, but different particle size, can also be added to the top layer (A).

Customary antiblocking agents (also referred to as pigments or particles) are inorganic and/or organic particles, for example calcium carbonate, amorphous silica, talc, magnesium carbonate, barium carbonate, calcium sulfate, barium sulfate, lithium phosphate, calcium phosphate, magnesium phosphate, aluminum oxide, lithium fluoride, calcium, barium, zinc or manganese salts of the dicarboxylic acids used, carbon black, titanium dioxide, kaolin or crosslinked polystyrene or acrylate particles. The particles can be added to the layer in the particular advantageous concentrations, for example as a glycolic dispersion during the polycondensation or via masterbatches in the course of the extrusion.

Particles which are preferred in accordance with the invention are synthetic, amorphous  $\text{SiO}_2$  particles in colloidal form. These particles are bound into the polymer matrix in an outstanding manner and generate only a few vacuoles (cavities). Vacuoles form at the particles in the course of the biaxial orientation, generally cause opacity and are therefore little suited to the present invention. To (synthetically) produce the  $\text{SiO}_2$  particles (also known as silica gel), sulfuric acid and sodium silicate are initially mixed together under controlled conditions to form hydrosol. This eventually forms a hard, transparent mass which is known as a hydrogel. After separation of the sodium sulfate formed as a by-product by a washing process, it can be dried and further processed. Control of the washing water pH and the drying

conditions can be used to vary the important physical parameters, for example pore volume, pore size and the size of the surface of the resulting silica gel. The desired particle size (for example the  $d_{50}$  value) and the  
5 desired particle size distribution (for example the SPAN98) are obtained by suitable grinding of the silica gel (for example mechanically or hydromechanically). Such particles can be obtained, for example, via Grace, Fuji, Degussa or Ineos.

10

It has been found to be particularly advantageous to use particles having an average particle diameter  $d_{50}$  of from 2.5 to 8  $\mu\text{m}$ , preferably from 3.0 to 7  $\mu\text{m}$  and more preferably from 3.5 to 6  $\mu\text{m}$ . When particles having a  
15 diameter which is below 2.5  $\mu\text{m}$  are used, there is no positive influence of the particles on the removal performance of the film from the tray. In this case, the film again tends to start to tear or continue to tear on removal from the tray, which is of course undesired.  
20 Particles having a diameter greater than 8  $\mu\text{m}$  generally cause filter problems.

In a preferred embodiment, the diameter of the particles in the heatsealable and peelable top layer (A) is greater  
25 than the thickness of this layer. It has been found to be advantageous to select a diameter/layer thickness ratio of at least 1.2, preferably at least 1.3 and more preferably at least 1.4. In these cases, there is a particularly positive influence of the particles on the  
30 removal performance of the film from the tray.

To provide the desired peeling properties, it has been found to be advantageous when the heatsealable and peelable top layer (A) comprises particles in a concen-  
35 tration of from 0.5 to 10 % by weight. The concentration

of the particles is preferably from 1.0 to 8.0 % by weight and more preferably from 1.5 to 6.0 % by weight. In contrast, when the top layer (A) of the film comprises particles and these are present in a concentration of less than 0.5 % by weight, there is no longer any positive influence on the removal performance of the film from the tray. In contrast, when the top layer (A) of the film comprises particles and these are present in a concentration of more than 10 % by weight, the opacity of the film becomes too high.

It has been found to be particularly advantageous to use particles in the heatsealable and peelable top layer (A) whose particle diameter distribution has a degree of scatter which is described by a SPAN98 of  $\leq 2.0$  (definition of SPAN98, see measurement method). Preference is given to a SPAN98 of  $\leq 1.9$  and particular preference to a SPAN98 of  $\leq 1.8$ . In contrast, when the top layer (A) of the film comprises particles whose SPAN98 is greater than 2.0, the optical properties and the sealing properties of the film deteriorate.

Moreover, it has been found to be advantageous to set the roughness of the heatsealable and peelable top layer (A) in such a way that its  $R_a$  value is greater than 60 nm. Preference is given to the roughness  $R_a$  being greater than 80 nm and it is more preferably greater than 100 nm; the upper limit of the roughness should not exceed 400 nm, preferably 350 nm, in particular 300 nm. This can be controlled via the selection of the particle diameters, their concentration and the variation of the layer thickness.

In order to further improve the processing performance of the film of the present invention, it is advantageous

likewise to incorporate particles into the base layer (B) in the case of a two-layer film structure (AB), or into the nonsealable top layer (C) in the case of a three-layer film structure (ABC), in which case the following  
5 conditions should be observed:

a) The particles should have an average particle diameter  $d_{50}$  (= median) of from 1.5 to 6  $\mu\text{m}$ . It has been found to be particularly appropriate to use  
10 particles having an average particle diameter  $d_{50}$  of from 2.0 to 5  $\mu\text{m}$  and more preferably from 2.5 to 4  $\mu\text{m}$ .

b) The particles should have a degree of scatter which is described by a SPAN98 of  $\leq 2.0$ . Preference is  
15 given to the SPAN98 being  $\leq 1.9$  and particular preference to the SPAN98 being  $\leq 1.8$ .

c) The particles should be present in a concentration of from 0.1 to 0.5 % by weight. The concentration of  
20 the particles is preferably from 0.12 to 0.4 % by weight and more preferably from 0.15 to 0.3 % by weight.

To achieve the aforementioned properties, in particular the optical properties of the sealable and peelable film, it has been found to be appropriate, in particular in the case of a three-layer film having ABC structure, to use a smaller amount of particles in the base layer (B) than  
30 in top layer (A). In the three-layer film of the type mentioned, the amount of particles in the base layer (B) should appropriately be between 0 and 2.0 % by weight, preferably between 0 and 1.5 % by weight, in particular between 0 and 1.0 % by weight. It has been found to be  
35 particularly appropriate only to incorporate those

particles into the base layer which get into the film via the same type of regrind (recyclate). The optical properties of the film, in particular the opacity of the film, are then particularly good.

5

Between the base layer and the top layers may optionally be disposed another intermediate layer. This may in turn consist of the polymers described for the base layer. In a particularly preferred embodiment, the intermediate layer consists of the polyesters used for the base layer. The intermediate layer may also comprise the customary additives described below. The thickness of the intermediate layer is generally greater than 0.3  $\mu\text{m}$  and is preferably in the range from 0.5 to 15  $\mu\text{m}$ , in particular in the range from 1.0 to 10  $\mu\text{m}$ , more preferably in the range from 1.0 to 5  $\mu\text{m}$ .

In the case of the two-layer and the particularly advantageous three-layer embodiment of the film according to the invention, the thickness of the top layer (A) is in the range from 0.7 and 2.5  $\mu\text{m}$ , preferably in the range from 0.8 and 2.2  $\mu\text{m}$  and more preferably in the range from 0.8 and 1.9  $\mu\text{m}$ . When the thickness of the top layer (A) is more than 2.5  $\mu\text{m}$ , the peeling force rises distinctly and is no longer within the inventive range. Moreover, the peeling performance of the film is impaired. In contrast, when the thickness of the top layer (A) is less than 0.5  $\mu\text{m}$ , the film is no longer heatsealable.

The thickness of the other, nonsealable top layer (C) may be the same as the top layer (A) or different; its thickness is generally between 0.5 and 5  $\mu\text{m}$ .

The total thickness of the inventive polyester film may vary within certain limits. It is from 3 to 200  $\mu\text{m}$ , in



particular from 4 to 150  $\mu\text{m}$ , preferably from 5 to 100  $\mu\text{m}$ , and the layer (B) has a proportion of preferably from 45 to 97 % of the total thickness.

5 The base layer and the other layers may additionally comprise customary additives such as stabilizers (UV, hydrolysis), flame-retardant substances or fillers. They are appropriately added to the polymer or the polymer mixture before the melting.

10

The present invention also provides a process for producing the film. To prepare the inventive heatsealable and peelable top layer (A), the particular polymers (polyester I, polyester II, polyester-incompatible  
15 polymer [anti-PET polymer], optionally further polymers [= for example masterbatch(es) for particles]) are appropriately fed directly to the extruder for the top layer (A). The materials can be extruded at from about 200 to 280 °C. From a process engineering point of view  
20 (mixing of the different components), it has been found to be particularly advantageous when the extrusion of the polymers for the top layer (A) is carried out using a twin-screw extruder having degassing means.

25 The polymers for the base layer (B) and for the further top layer (C) which may possibly be present and optionally the intermediate layer are appropriately fed to the (coextrusion) system via further extruders. The melts are shaped to flat melt films in a multilayer die  
30 and layered on top of one another. Subsequently, the multilayer film is drawn off with the aid of a chill roll and optionally further rolls and solidified.

The biaxial stretching of the film is generally carried  
35 out sequentially. Simultaneous stretching of the film is

also possible, but is not necessary. In the sequential stretching, preference is given to stretching first in longitudinal direction (i.e. in machine direction) and then in transverse direction (i.e. at right angles to machine direction). The stretching in the longitudinal direction can be carried out with the aid of two rolls rotating at different rates in accordance with the desired stretching ratio. For transverse stretching, an appropriate tenter frame is generally used.

10

The temperature at which the stretching is carried out can be varied within a relatively wide range and depends on the desired properties of the film. In general, the stretching is carried out in longitudinal direction (machine direction orientation = MDO) in a temperature range of from 60 to 130 °C (heating temperatures from 60 to 130 °C), and in transverse direction (transverse direction orientation = TDO) in a temperature range from 90 °C (beginning of the stretching) to 140 °C (end of the stretching). The longitudinal stretching ratio is in the range from 2.0:1 to 5.5:1, preferably from 2.3:1 to 5.0:1. The transverse stretching ratio is generally in the range from 2.4:1 to 5.0:1, preferably from 2.6:1 to 4.5:1.

25

The preferred temperature range at which the biaxial stretching is carried out in the longitudinal stretching (MDO) is from 60 to 120 °C. The heating temperatures of the film in the longitudinal stretching are in the range from 60 to 115 °C. In the transverse stretching (TDO), the temperatures of the film are in the range from 90 °C (beginning of the stretching) to 140 °C (end of the stretching). The longitudinal stretching ratio in this preferred temperature range is in the range from 2.0:1 to 5.0:1, preferably from 2.3:1 to 4.8:1. The transverse

35

stretching ratio is generally in the range from 2.4:1 to 5.0:1, preferably from 2.6:1 to 4.5:1.

5 The particularly preferred temperature range in which the  
biaxial stretching is carried out in the case of the  
longitudinal stretching (MDO) is from 60 to 110 °C. The  
heating temperatures of the film in the longitudinal  
stretching are in the range from 60 to 105 °C. In the  
transverse stretching (TDO), the temperatures of the film  
10 are in the range from 90 °C (beginning of the stretching)  
to 140 °C (end of the stretching). The longitudinal  
stretching ratio in this preferred temperature range is  
in the range from 2.0:1 to 4.8:1, preferably from 2.3:1  
to 4.6:1. The transverse stretching ratio is generally in  
15 the range from 2.4:1 to 5.0:1, preferably from 2.6:1 to  
4.5:1.

The preferred and especially the particularly preferred  
temperatures in the MDO particularly effectively take  
20 into account the adherent behavior of top layer (A) to  
rolls (metallic, ceramic or particularly coated roll  
surfaces).

Before the transverse stretching, one or both surface(s)  
25 of the film can be coated inline by the processes known  
per se. The inline coating may lead, for example, to  
improved adhesion between a metal layer or a printing ink  
and the film, to an improvement in the antistatic  
performance, in the processing performance or else to  
30 further improvement of barrier properties of the film.  
The latter is contained, for example, by applying barrier  
coatings such as EVOH, PVOH or the like. In that case,  
preference is given to applying such layers to the  
nonsealable surface, for example the surface (C) of the  
35 film.

In the subsequent heat-setting, the film is kept at a temperature of from 150 to 250 °C over a period of from about 0.1 to 10 s. Subsequently, the film is wound up in a customary manner.

5

The gloss of the film surface (B) in the case of a two-layer film, or the gloss of the film surface (C) in the case of a three-layer film, is greater than 100 (measured to DIN 67530 based on ASTM-D 523-78 and ISO 2813 with  
10 angle of incidence 20 °). In a preferred embodiment, the gloss of these sides is more than 110 and in a particularly preferred embodiment more than 120. These film surfaces are therefore suitable in particular for a further functional coating, for printing or for  
15 metallization.

The opacity of the film is less than 20. In a preferred embodiment, the opacity of the film is less than 16 and in a particularly preferred embodiment less than 12.

20

A further advantage of the invention is that the production costs of the film according to the invention are not substantially above those of a film made of standard polyester. In addition, it is guaranteed that, in the  
25 course of the production of the film, offcut material which arises intrinsically in the operation of the film production can be reused for the film production as regrind in an amount of up to 60 % by weight, preferably from 5 to 50 % by weight, based in each case on the total  
30 weight of the film, without the physical properties of the film being significantly adversely affected.

The film according to the invention is outstandingly suitable for packaging foods and other consumable goods,  
35 in particular in the packaging of foods and other

consumable goods in trays in which peelable polyester films are used for opening the packaging.

The table which follows (table 1) once again summarizes  
5 the most important inventive film properties.

Table 1

	<b>Top layer (A)</b>	Inventive range	Preferred	Particularly preferred	Unit	Measurement method
5	Proportion of units in the inventive polyester which are based on aromatic dicarboxylic acids	20 to 95	30 to 900	50 to 88	mol%	
10	Proportion of units in the inventive polyester which are based on aliphatic dicarboxylic acids	5 to 80	10 to 70	12 to 50	mol%	
	Anti-PET polymer	3 to 20	5 to 18	7 to 15	% by wt.	
	Polyester I	10 to 60	15 to 55	20 to 50	% by wt.	
15	Polyester II	20 to 80	25 to 80	30 to 80	% by wt.	
	Particle diameter $d_{50}$	2.5 to 8	3.0 to 7	3.5 to 6	$\mu\text{m}$	internal
	Filler concentration	0.5 to 10.0	1.0 to 8.0	1.5 to 6.0	%	internal
	Thickness of the top layer A	1.7 to 6.0	2.0 to 5.0	2.5 to 4.0	$\mu\text{m}$	
20	Particle diameter/layer thickness ratio	$\geq 1.2$	$\geq 1.3$	$\geq 1.4$		

**Properties**

Thickness of the film	3 to 200	4 to 150	5 to 100	µm	
Minimum sealing temperature of TL(A) with respect to CPET trays	165	160	155	°C	internal
Seal seam strength of TL (A) with respect to CPET trays	1.5 to 8	2.0 to 7.5	2.5 to 7.0	N/15 mm	internal
Gloss of the top layers A and C	> 70 and > 100	> 75 and > 110	> 80 and > 120		DIN 67530
Opacity of the film	< 20	< 16	< 12	%	ASTM D 1003-52

10 TL: Top layer, >/=: greater than/equal to

To characterize the raw materials and the films, the following measurement methods were used for the purposes of the present invention:

5    **Measurement of the average diameter  $d_{50}$**

The determination of the average diameter  $d_{50}$  was carried out by means of a laser on a Malvern Master Sizer by means of laser scanning (other measuring instruments are, for example, Horiba LA 500 or Sympatec Helos, which use the same measuring principle). To this end, the samples were introduced together with water into a cuvette and this was then placed in the measuring instrument. The dispersion is scanned by means of a laser and the signal is used to determine the particle size distribution by comparison with a calibration curve. The particle size distribution is characterized by two parameters, the median value  $d_{50}$  (= measure of the position of the average value) and the degree of scatter, known as the SPAN98 (=measure of the scatter of the particle diameter). The measuring procedure is automatic and also includes the mathematical determination of the  $d_{50}$  value. The  $d_{50}$  value is determined by definition from the (relative) cumulative curve of the particle size distribution: the point at which the 50 % ordinate value cuts the cumulative curve provides the desired  $d_{50}$  value (also known as median) on the abscissa axis.

**Measurement of SPAN98**

The determination of the degree of scatter, the SPAN98, was carried out with the same measuring instrument as described above in the determination of the average diameter  $d_{50}$ . The SPAN98 is defined as follows:



$$SPAN\ 98 = \frac{d_{98} - d_{10}}{d_{50}}$$

The basis of the determination of  $d_{98}$  and  $d_{10}$  is again the (relative) cumulative curve of the particle size distribution (see above "measurement of the average diameter  $d_{50}$ "). The point at which the 98 % ordinate value cuts the cumulative curve provides the desired  $d_{98}$  value directly on the abscissa axis and the point at which the 10 % ordinate value of the cumulative curve cuts the curve provides the desired  $d_{10}$  value on the abscissa axis.

#### 10 **SV value**

The SV value of the polymer was determined by the measurement of the relative viscosity ( $\eta_{rel}$ ) of a 1 % solution in dichloroacetic acid in an Ubbelohde viscometer at 25 °C. The SV value is defined as follows:

15  $SV = (\eta_{rel} - 1) \cdot 1000.$

#### **Glass transition temperatures $T_g$**

The glass transition temperature  $T_g$  was determined using film samples with the aid of DSC (differential scanning calorimetry). The instrument used was a Perkin-Elmer DSC 1090. The heating rate was 20 K/min and the sample weight approx. 12 mg. In order to eliminate the thermal history, the samples were initially preheated to 300 °C, kept at this temperature for 5 minutes and then subsequently quenched with liquid nitrogen. The thermogram was used to find the temperature for the glass transition  $T_g$  as the temperature at half of the step height.

**Seal seam strength**

To determine the seal seam strength, a film strip (100 mm long × 15 mm wide) is placed on an appropriate strip of the CPET tray and sealed at the set temperature of  
5 > 140 °C, a sealing time of 0.5 s and a sealing pressure of 3 bar (Brugger HSG/ET sealing unit, sealing jaw heated on both sides). In accordance with **figure 2**, the sealed strips are clamped into the tensile testing machine (for example Zwick) and the 180 ° seal seam strength, i.e. the  
10 force required to separate the test strips, was determined at a removal rate of 200 mm/min. The seal seam strength is quoted in N per 15 mm of film strip (e.g. 3 N/15 mm).

**15 Determination of the minimum sealing temperature**

The Brugger HSG/ET sealing unit as described above for the measurement of the seal seam strength is used to produce heatsealed samples (seal seam 15 mm × 100 mm), and the film is sealed at different temperatures with the  
20 aid of two heated sealing jaws at a sealing pressure of 3 bar and a sealing time of 0.5 s. The 180 ° seal seam strength was measured as for the determination of the seal seam strength. The minimum sealing temperature is the temperature at which a seal seam strength of at least  
25 1.7 N/15 mm is attained.

**Roughness**

The roughness  $R_a$  of the film was determined to DIN 4768 at a cutoff of 0.25 mm. It was not measured on a glass  
30 plate, but rather in a ring. In the ring method, the film is clamped into a ring, so that neither of the two surfaces touches a third surface (for example glass).

**Opacity**

The opacity according to Hölz was determined to ASTM-D 1003-52.

5 **Gloss**

The gloss of the film was determined to DIN 67530. The reflector value was measured as a characteristic optical parameter for the surface of a film. Based on the standards ASTM-D 523-78 and ISO 2813, the angle of incidence was set to 20 °. A light beam hits the flat test surface at the angle of incidence set and is reflected or scattered by it. The lightbeams incident on the photoelectronic detector are displayed as a proportional electrical quantity. The measurement is dimensionless and has to be quoted together with the angle of incidence.

**Tensile strain at break**

20 The tensile strain at break of the film was determined to DIN 53455. The extension rate is 1 %/min; 23 °C; 50 % relative humidity.

**Modulus of elasticity**

25 The modulus of elasticity of the film was determined to DIN 53457. The extension rate is 1 %/min; 23 °C; 50 % relative humidity.

**Shrinkage**

30 The gloss of the film was [lacuna] to DIN 40634. The testing conditions are 150 °C, 15 min.

The invention is illustrated hereinbelow with the aid of examples.

**Example 1**

Chips of polyethylene terephthalate were fed to the extruder for the base layer (B). Chips of polyethylene terephthalate and particles were likewise fed to the  
5 extruder (twin-screw extruder) for the nonsealable top layer (C). In accordance with the process conditions listed in the table below, the raw materials were melted and homogenized in the two respective extruders.

10 In addition, a mixture consisting of polyester I, polyester II and anti-PET polymer was prepared for the heatsealable and peelable top layer (A). In table 2, the particular proportions of the dicarboxylic acids and glycols present in the two polyesters I and II in mol%  
15 and the particular proportions of the components present in the mixture in % by weight are specified. The mixture was fed to the twin-screw extruder with degassing for the sealable and peelable top layer (A). In accordance with the process conditions detailed in the table below, the  
20 raw materials were melted and homogenized in the twin-screw extruder.

By coextrusion in a three-layer die, the three melt streams were then layered on top of one another and  
25 ejected via the die lip. The resulting melt film was cooled and a transparent, three-layer film having ABC structure was subsequently produced in a total thickness of 25  $\mu\text{m}$  by a stepwise orientation in the longitudinal and transverse direction. The thicknesses of the two top  
30 layers are each 2.0  $\mu\text{m}$  (cf. also table 2).

Top layer (A), mixture of:

25.0 % by weight of polyester I (= copolymer of  
78 mol% of ethylene terephthalate, 22

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5 mol% of ethylene isophthalate) having  
an SV value of 850. The glass  
transition temperature of polyester I  
is approx. 75 °C. Polyester I  
5 additionally contains 5.0 % by weight  
of <sup>®</sup>Sylysia 430 (synthetic SiO<sub>2</sub>, Fuji,  
Japan) having a particle diameter of  
d<sub>50</sub> = 3.4 µm and a SPAN98 of 1.7. The  
10 ratio of particle diameter d<sub>50</sub> to top  
layer thickness d<sub>(A)</sub> is 1.7:1 (cf.  
table 2).

60 % by weight of polyester II (= copolymer  
containing 40 mol% of ethylene  
azelate, 50 mol% of ethylene  
15 terephthalate, 10 mol% of  
ethylene isophthalate) having an  
SV value of 1000. The glass  
transition temperature of  
polyester II is approx. 0 °C.

20 15 % by weight of anti-PET polymer (= COC,  
<sup>®</sup>Topas 8007, Ticona, Frankfurt;  
having a T<sub>g</sub> of approx. 75 °C)

Base layer (B):

25 100 % by weight of polyethylene terephthalate  
having an SV value of 800

Top layer (C), mixture of:

85 % by weight of polyethylene terephthalate  
30 having an SV value of 800

15 % by weight of a masterbatch of 99 % by  
weight of polyethylene  
terephthalate (SV value of 800)  
and 1.0 % by weight of Sylobloc

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44 H (synthetic SiO<sub>2</sub>, Grace,  
Worms), d<sub>50</sub> = 2.5 µm, SPAN98 =  
1.9

5 The production conditions in the individual process steps  
were:

10	Extrusion	Temperatures	A layer:	2e+08	°C
			B layer:		°C
			C layer:		°C
			Temperature of the takeoff roll	20	°C
	Longitudinal stretching	Heating temperature		70-90	°C
			Stretching temperature	95	°C
			Longitudinal stretching ratio	38	
	Transverse stretching	Heating temperature		100	°C
			Stretching temperature	135	°C
			Transverse stretching ratio	40	
15	Setting	Temperature		230	°C
		Time		3	s

20 Table 3 shows the properties of the film. According to  
measurements (column 2), the minimum sealing temperature  
of the film with respect to CPET trays is 148 °C. The  
film was sealed to the CPET trays at 160, 180 and 200 °C  
(sealing pressure 4 bar, sealing time 0.5 s).  
25 Subsequently, strips of the bond of inventive film and  
CPET tray were pulled apart by means of a tensile strain

tester in accordance with the aforementioned measurement method (cf. figure 2). For all sealing temperatures, the films exhibited the desired peeling off from the tray according to figure 3b. The seal seam strengths measured  
5 are listed in column 3. For all sealing temperatures, peelable films were obtained. The seal seam strengths at approx. 5 N/15 mm are within the medium range, i.e. the films can be removed from the tray without great force being applied. In addition, the film had the required  
10 good optical properties, exhibited the desired handling and the desired processing performance.

### Example 2

In comparison to example 1, the composition of the  
15 mixture for the sealable top layer (A) was changed. The composition of the individual components remained unchanged in comparison to example 1. The mixture now consists of the following raw material proportions:

- polyester I = 20 % by weight,
- 20 • polyester II = 70 % by weight and
- anti-PET polymer = 10 % by weight.

As a consequence of the higher proportion of polyester II in the mixture, the process parameters were modified in the longitudinal stretching. The new conditions for the  
25 longitudinal stretching are listed in the table below.

Longitudinal stretching	Heating temperature	70-92	°C
	Stretching temperature	94	°C
30	Longitudinal stretching ratio	3.7	

The minimum sealing temperature of the film with respect to the CPET trays is now 146 °C. For all sealing temperatures, the films exhibited the desired peeling off from the tray according to figure 3b. The seal seam strengths measured are listed in column 3. For all sealing temperatures, peelable films were again obtained. The seal seam strengths of the films according to the invention are lower than in example 1. They are within the medium range, so that the film can be removed from the tray without substantial force being applied. The handling and the processing performance of the film was as in example 1.

**Example 3**

In comparison to example 2, the top layer thickness of the sealable layer (A) was raised to 3.0 µm. The diameter of the filler was raised to 4.0 µm. The minimum sealing temperature of the film with respect to CPET trays is now 144 °C. For all sealing temperatures, the films exhibited the desired peeling off from the tray according to figure 3b. The seal seam strengths measured are listed in column 3. For all sealing temperatures, peelable films were again obtained. The seal seam strengths of the inventive films are somewhat higher than in example 2. A somewhat lower opacity of the film was measured; the handling and the processing performance of the film was as in example 1.

**Example 4**

In comparison to example 3, the composition of polyester II for the sealable top layer (A) was changed. In comparison to example 2, the top layer thickness of the sealable layer (A) was raised to 3.4 µm. The diameter of the filler was raised to 5.0 µm. The mixture used in top



layer (A) now consists of the following raw material proportions:

- 15 % by weight of polyester I, identical to example 1
- 5 80 % by weight of polyester II, \*Vitel1912, (polyester, Bostik-Findley, USA; contains the dicarboxylic acid constituents azelaic acid, sebacic acid, terephthalic acid, isophthalic acid and further dicarboxylic acids approximately in the molar ratio 40/1/45/10/4, and, as the diol component, at least 60 mol% of ethylene glycol). The glass transition temperature of polyester
- 10
- 15 II is approx. -1 °C.
- 5 % by weight of COC (\*Topas 8007, Ticona, Frankfurt; an ethylene/norbornene COC having a  $T_g$  of approx. 75 °C)

20

- The process parameters in the longitudinal stretching corresponded to those in example 3. The minimum sealing temperature of the film produced in accordance with the invention with respect to the CPET trays is now 142 °C.
- 25 For all sealing temperatures, the films exhibited the desired peeling off from the tray according to figure 3b. The seal seam strengths measured are listed in column 3. For all sealing temperatures, peelable films were again obtained. They are within the medium range, so that the
- 30 film can be removed from the tray without substantial force being applied. The handling and the processing performance of the film was as in example 1.

**Comparative example 1**

In comparison to example 1, the composition of the sealable layer (A) was changed. In the top layer (A), only the polyester I based on aromatic acids was used:

5

100.0 % by weight of polyester I (= copolymer of 78 mol% of ethylene terephthalate and 22 mol% of ethylene isophthalate) having an SV value of 850. The glass transition temperature of polyester I is approx. 75 °C. In addition, polyester I contains 5.0 % of ®Sylysia 430

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15

The production conditions in the individual process stages were adapted in the longitudinal stretching to the glass transition temperature of the top layer raw material:

20

Longitudinal stretching	Heating temperature	70-115 °C
	Stretching temperature	120 °C
	Longitudinal stretching ratio	4.0

25

Table 3 shows the properties of the film. Although the film is highly pigmented and the pigments constitute weak points in the sealing layer, a peelable film was not obtained for any of the specified sealing temperatures.

30

On removal of the film from the tray, the film started to tear immediately and exhibited a force-path diagram according to figure 3b. The film exhibits weldable

behavior and is thus unsuitable for the achievement of the object specified.

**Comparative example 2**

5 Example 5 from EP-A 0 035 835 was reproduced. Table 3 shows the properties of the film. A peelable film was not obtained for any of the specified sealing temperatures. On removal of the film from the tray, the film started to  
10 tear immediately and exhibited a force-path diagram according to figure 3b. The film exhibits weldable behavior and is thus unsuitable for the achievement of the object specified.

**Comparative example 3**

15 Example 1 from EP-A 0 379190 was reproduced. Table 3 shows the properties of the film. A peelable film was not obtained for any of the specified sealing temperatures. On removal of the film from the tray, the film started to tear immediately and exhibited a force-path diagram  
20 according to figure 3b. The film exhibits weldable behavior and is thus unsuitable for the achievement of the object specified.

**Comparative example 4**

25 Example 22 from EP-A 0 379190 was reproduced. Table 3 shows the properties of the film. A peelable film was not obtained for any of the specified sealing temperatures. On removal of the film from the tray, the film started to tear immediately and exhibited a force-path diagram  
30 according to figure 3b. The film exhibits weldable behavior and is thus unsuitable for the achievement of the object specified.

Table 2

	Polyester I composition				Polyester II composition								Anti-PET polymer	PI/PII/ anti-PET polymer ratios	Glass transition temperatures PI/PII/ anti-PET polymer	Film structure	Film thickness	Top layer thicknesses		Particles			d <sub>50</sub> / d <sub>10</sub> ratio
	TA	IA	EG	NG	AzA	SeA	AdA	TA	IA	EG	BD	FA	COC				(A)	(C)	Dia-meter	SPAN 98	Conc		
	mol%				mol%									% by wt	° C		μm	μm	μm	-	%		
5	E 1	78	22	100	40			50	10	100			100	25/60/15	75/0/75	ABC	25	2	2	3.4	1.8	1.25	1.7
	a 2	78	22	100	40			50	10	100			100	20/70/10	75/0/75	ABC	25	2.5	2.5	3.4	1.8	1.00	1.36
	m 3	78	22	100	40			50	10	100			100	20/70/10	75/0/75	ABC	25	3	3	4	1.8	1.00	1.33
10	e 4	78	22	100	40	1		45	10	>60		4	100	15/80/5	75/-/175	ABC	25	3.4	3.4	5	1.8	0.75	1.47
	C 1	78	22	100	-	-	-	-	-	-	-	-	-	100/0/0/	75	ABC	25	1	1	3.4	1.8	5	3.4
	E 2	82	18	100	-	-	-	-	-	-	-	-	-	100/0/0/	75	AB	20	2.98	-	1.5+5	-	0.3	1.68
15	a 3	-	-	-		10		90		100				0/100/0/	approx. 50	AB	17.2	4.1	-	-	-	-	-
	m 4	100	-	85	15.4	31.5	2.4	65	1.1	95.4	4.6		-	50/50/0/	approx. 20	AB	11.5	2.5	-	2	-	0.25	0.8

TA terephthalate, IA isophthalate, EG ethylene, BD butane, NG neopentyl

AZA azelate, SeA sebacate, AdA adipate, FA further dicarboxylic acids and glycols

Table 3

	Minimum sealing temperature	Seal seam strength with respect to CPET trays			Peeling test (= peeling performance)	Opacity	Gloss		Roughnesses	
		160 °C	180 °C	200 °C			A side	C side	A side	C side
E x a m p l e s	°C					%			µm	
	1	148	3.5	5.2	7.4	13	90	133	190	60
	2	146	3.2	3.5	6.3	11	90	133	188	60
	3	144	4.5	5.4	7.2	10	102	140	170	60
	4	142	4.8	4.7	6.1	8	110	130	156	60
C e x a m p l e s	1	105	3.5	5	8	23	55	130	310	60
	2	109	4.2	5.5	8.1	13	110	190	69	25
	3	112	2	4	6	4	150	190	33	20
	4	110	3	4	5	1.5	130	190	120	22

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	Peeling test:	At all sealing temperatures, film is peeled from the tray without the film starting to tear or continuing to tear. Impeccable, smooth, clean peeling of the film from the tray, even in the upper temperature range at high seal seam strength.
		At all sealing temperatures, film starts to tear on removal from the tray.